Data Analysis Techniques Using Hydrocarbon and Carbonyl Compounds

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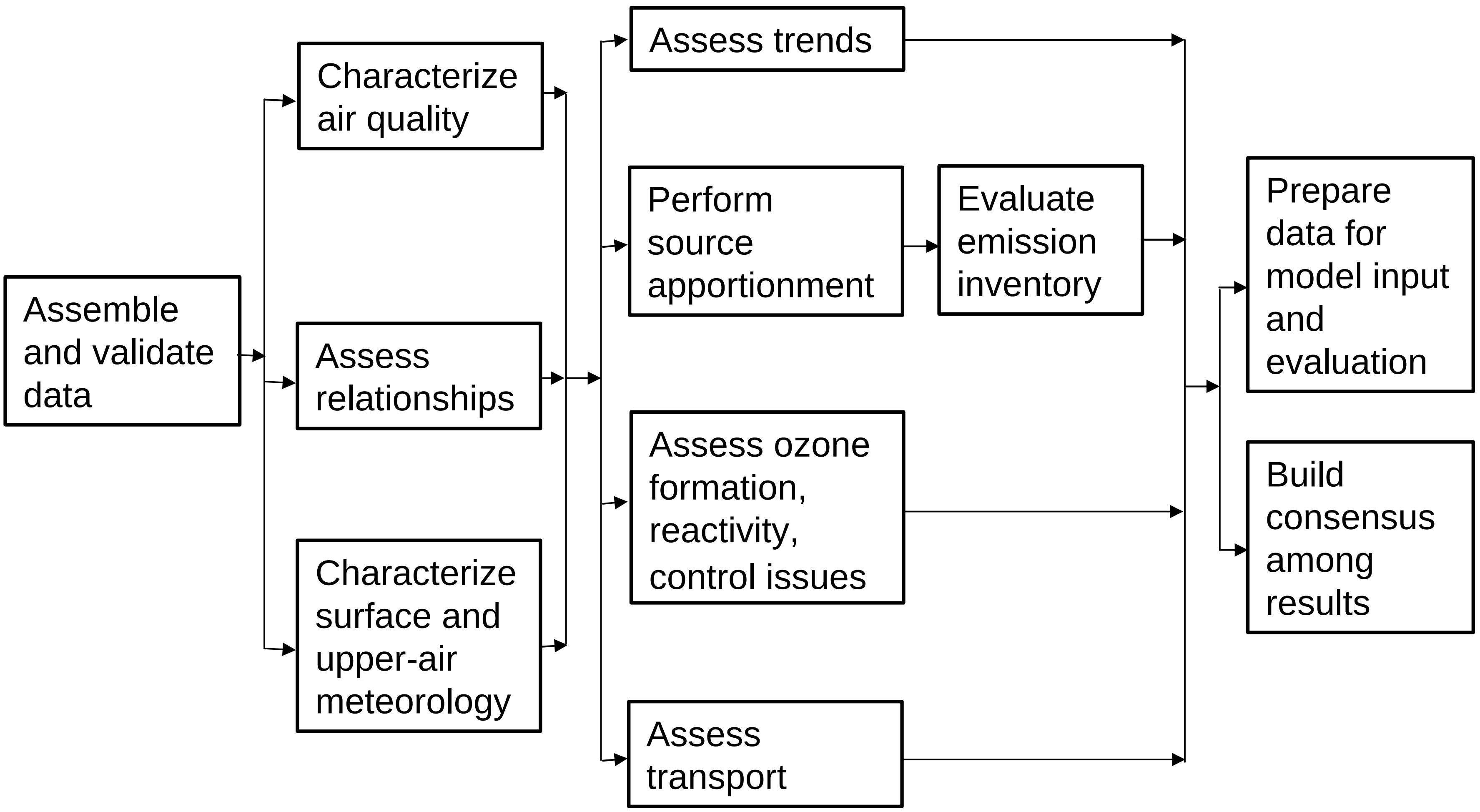
Introduction

- The purpose of a description and analysis of the validated PAMS data includes assessing ozone formation, transport, and control strategies. To aid in these assessments, useful analyses include the statistical description of the data including the spatial and temporal variations.
- The process of describing and displaying the PAMS data includes:
  - retrieving data
  - reviewing the data validity and completeness
  - defining objectives of the analyses
  - applying several techniques and approaches in order to obtain consensus among results
- The intent of this section is to provide examples of the prescribed analyses presented in the Introduction as well as other analyses that could be beneficial in answering questions about a region's air quality.

Example Flow of Analyses



Example flowchart of PAMS data analyses to be applied in order to meet PAMS objectives.

Available Tools and Methods

Tools and methods available to investigate the PAMS data include the following:

- Statistical software and related tools (e.g., AMDAS from *http://www.environ.org/amdas*).
- PAMS VOC data validation and data display tool (e.g., VOCDat from *ftp://ftp.sonomatech.com/public/vocdat/*). This tool is described in more detail in the Data Validation section.
- Spreadsheets and graphical packages

Example VOC Data Exploration Tool (1 of 3)

- AMDAS is an add-on module for S-PLUS (a statistical package.
- AMDAS allows the analyst to perform the types of analyses most frequently used to characterize and evaluate ambient data including statistical summaries, time series, scatter plot matrices, regression analysis, side-by-side box plots, diurnal profiles, fingerprint plots, and pollution roses.

Ambient Monitoring Data Analysis System (AMDAS)

Stoeckenius, 1999

AMDAS requires S-Plus for Windows version 4.5 or above

Example VOC Data Exploration Tool (2 of 3)

- AMDAS has four main menu groups: Data, Plots, Summaries, and Utilities.
- The regular S-PLUS menu options and object explorer are also available for use.
- Under the data menu the following options are available: import AMP-370 file (old AIRS format) and R-2 file (new AIRS format), calculate daily statistics of 1-hr and 3-hr data, convert 1-hr to 3-hr data, create weight percent data, merge datasets, and create composite variables (such as ratios).
- The summary menu features the following options: typical ranges table for PAMS VOC data, a rank table showing VOC species rankings across sites or years of data, and tables of summary statistics.
- The utilities menu allows the user to set flags of individual data points, edit user defined flags, browse the data, modify data dictionary contents, and add AIRS codes to the dictionary.

Example VOC Data Exploration Tool (3 of 3)

Under the plot menu, the following options are available:

- scatter plot matrices provide a convenient means of identifying relationships among variables
- diurnal profiles compare the mean daily cycle of several different variables between different monitoring sites, different years, or weekend vs. weekday
- box plots present compact summaries of the statistical distributions of variables
- time series show raw data values against a user-defined time axis
- simple regression present results of linear least-squares regression
- multiple regression presents results for up to five independent variables
- overlaid simple regression allows two simple regression plots to be overlaid
- trends in user-selected annual summary statistics
- fingerprint plots show composition of a sample
- pollution rose plots show concentration by wind direction

Assemble and Validate Data (1 of 2)

- After obtaining the data, the analyst should also obtain as much supplemental information regarding the data quality as possible. This information is critical to the analyst because it provides needed insight on the accuracy and precision of the data, the potential for contamination, potential impact of nearby sources, and potential issues with the database because of the use of a particular analytical technique.
- Information includes:
  - Sample collection specifications and special features such as the use of a Nafion dryer to handle moisture in VOC samples (e.g., Nafion dryers are known to remove oxygenated species and some biogenics).
  - Sampling location description (e.g., What are nearby sources? topography?)
  - Audit, blank collection, collocated sampler descriptions (e.g., assess accuracy, precision)
  - Sample analysis and instrument calibration descriptions (e.g., assess methodology)
  - Example calculations of concentrations and any data conversions

Assemble and Validate Data (2 of 2)

- Information includes (continued):
  - Laboratory quality control/quality assurance descriptions
  - Reported units, site, date, sample start and end times, specification of daylight or standard time
  - Species detection limits and lower quantifiable limits
  - Treatment of missing data and data below detection
- Next, review the validation status of the data and perform additional validation steps if necessary. Data validation is critical because serious errors in data analysis and modeling results can be caused by erroneous individual data values.

Defining Analysis Objectives (1 of 2)

Decision matrix to be used to identify example activities that will help the analyst address science/technical questions and objectives. To use the matrix, find your technical objective at the top left. Follow this line across to see which example activities will be useful to meet the objective. For each of these activities, look down the column to see which data and data analysis tools are useful for the activity (on the next page).

	APPROACHES, TOOLS																	
SCIENCE/TECHNICAL QUESTIONS/OBJECTIVES	Describe & display spatial & temporal O3 distributions	Describe & display spatial & temporal precursor distributions	Perform climatological, synoptic analyses	Compare surface and upper-air AQ data	Estimate pollutant fluxes	Describe & display met characteristics	Perform case study of met & AQ at strategic sites	Perform trajectory analyses	Compare ambient & emissions ratios, fractions, composition	Interpretative & case study analyses for model input, evaluation	Develop conceptual model	Perform VOC receptor modeling	Apply Smog Production Algorithms	Apply emissions-based models	Develop statistical relationships among pollutants	Characterize reaction chemistry	2D and 3D displays and evaluations	Analysis of model simulation results
O3 concentration distributions	x			x														
Precursor concentration distributions		x		x														
Fluxes into and within region				x	x													x
Meteorological processes				x		x	x											
Climatological patterns			x															
Evaluate Emissions Inventories									x			x						x
Source attribution								x				x						x
VOC sources (natural vs. anthropogenic)												x						x
VOC and/or NOx reduction influence on O3													x	x	x	x		x
Data for model initial, boundary conditions										x								
Air qulaity model evaluation										x	x					x	x	x
Meteorolgical model evaluation										x	x						x	x
Met & AQ phenomena to be reproduced by models											x					x	x	x
Contribution of subregions, outside OTR, carryover											x							x

Defining Analysis Objectives (2 of 2)

Decision matrix continued. This portion of the matrix indicates the useful measurements and tools associated with each example activity.

	APPROACHES, TOOLS																	
	Describe & display spatial & temporal O3 distributions	Describe & display spatial & temporal precursor distributions	Perform climatological, synoptic analyses	Compare surface and upper-air AQ data	Estimate pollutant fluxes	Describe & display met characteristics	Perform case study of met & AQ at strategic sites	Perform trajectory analyses	Compare ambient & emissions ratios, fractions, composition	Interpretative & case study analyses for model input, evaluation	Develop conceptual model	Perform VOC receptor modeling	Apply Smog Production Algorithms	Apply emissions-based models	Develop statistical relationships among pollutants	Characterize reaction chemistry	2D and 3D displays and evaluations	Analysis of model simulation results
MEASUREMENTS																		
Surface																		
O3	x			x			x			x	x		x	x	x	x	x	x
NO, NOx or NOy		x		x			x		x	x	x		x	x	x	x	x	x
NMHC, carbonyls		x		x			x		x	x	x	x	x	x	x	x	x	x
Meteorology			x				x	x	x	x	x			x	x		x	x
Upper-air																		
Meteorology			x		x	x	x			x	x			x	x		x	x
Non-PAMS																		
CO									x	x		x	x		x			x
Emission Inventory									x					x				x
UAM Model Output																		x
Aloft Air Quality	x	x		x	x	x				x	x				x	x	x	x
USEFUL TOOLS																		
AMDAS	✓	✓	✓	✓	✓					✓	✓			✓		✓		
VOCDat		✓								✓	✓					✓		
Other Statistical Methods	✓	✓	✓	✓	✓	✓			✓	✓	✓			✓		✓		
Trajectory Methods				✓	✓			✓		✓		✓		✓			✓	
Factor, cluster analyses						✓						✓			✓	✓	✓	
Advanced factor analyses (e.g., PMF, UNMIX)															✓	✓	✓	
CMB																✓	✓	
SPECIATE														✓	✓	✓	✓	✓

Spatial and Temporal Characteristics

- In general, a data analysis project proceeds from a complete understanding of the data, to a thorough validation of the data, to data analysis tasks.
- Initial data analysis tasks usually display and describe the data, while later analysis tasks are usually more complex and interpretive.
- No one analysis should determine an action or form the basis of a conclusion. Analysts should apply many different techniques and approaches and strive to obtain consensus among results.

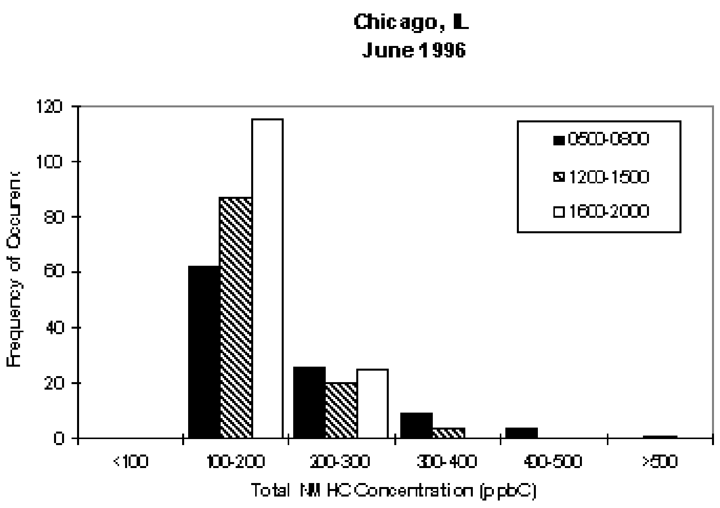
Summary Statistics (1 of 2)

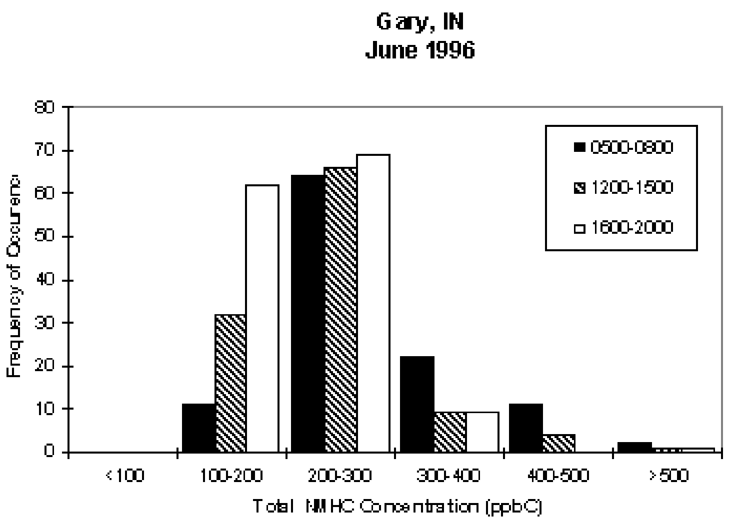
Site	Site Type	NMHC (ppbC)	Isoprene %	Toluene/ Benzene
Corbin, VA	1	59	13	2.2
Lums Pond, DE	1/4	30	3.7	1.7
Arendtsville, PA	Rural	23	4.1	1.9
Fort Meade, MD	1/3	98	3.2	2.4
New Brunswick, NJ	1/4	102	2.9	2.9
N. Carolina	Mix	112	3.3	3.4
Camden, NJ	2	109	0.65	2.8
Essex, MD	2	89	1.9	3.3
Lake Clifton, MD	2	115	0.55	2.5
McMillan Reservoir, DC	2	173	1.4	2.9
Philadelphia, PA	2	210	0.93	2.8
Aldino, MD	3	73	1.0	2.6
Rider College, NJ	3	45	3.4	2.6

- It is helpful to have an overall understanding of the database before proceeding to a more detailed analysis. This example summarizes the median total NMHC concentration, isoprene %, and toluene:benzene ratio at each site in the Mid-Atlantic region in 1997 (Main et al., 1999).
- NMHC values are an indicator of emission source strength impacting the site; the concentrations were generally higher at the Type 2 sites than at the other site types. Isoprene is the only tracer for biogenic emissions at these sites. We might expect the isoprene contribution to be higher for more rural sites. The toluene:benzene ratio is an indicator of source strength and the proximity of fresh emissions. Toluene reacts away faster in the atmosphere than benzene and , thus, a lower ratio can be an indication of a more aged air mass. We would expect higher ratios for the Type 2 sites.

Summary Statistics (2 of 2)

- Histograms are another useful tool for examining the distribution of concentrations in a database.
- This example shows the number of samples with NMHC concentrations in selected ranges by time of day and site for June 1996 data.
- The Gary site NMHC concentrations were typically higher than the Chicago site data.
- Also note that the highest concentrations typically occurred in the morning and the lowest concentrations typically occurred in the afternoon. This finding is consistent with meteorology (lower mixing heights in the morning, higher in the afternoon) and emissions patterns.





Ten Most Abundant Hydrocarbons (1 of 2)

- Determine the most abundant species based on median concentration to potentially shorten the number of species to be considered in subsequent analyses.
- How does the list compare among sites and site types? Isoprene is typically abundant at PAMS non-Type 2 sites as illustrated in this example.

Lums Pond, DE 1997
PAMS Type 1/4

Concentration
Ethane
Toluene
i-Pentane
Propane
n-Butane
n-Pentane
m-&p-Xylenes
Benzene
Isoprene
i-Butene

Main et al., 1999

Determine the median value of all species concentrations over the desired time frame (e.g., sample time, month, year) and then sort the median values.

Ten Most Abundant Hydrocarbons (2 of 2)

- Inspect the concentration lists. Are there any surprises? At the example PAMS Type 2 site (urban) shown here, biogenic isoprene is not among the most abundant species as was observed at Lums Pond.
- How has the list changed from year to year? For example, benzene used to be more abundant. Benzene's decrease in abundance reflects targeted control measures on gasoline benzene content.

McMillan Reservoir (Washington, D.C.) 1997
PAMS Type 2

Concentration
Ethane
Toluene
i-Pentane
Propane
n-Butane
n-Pentane
Ethene
m-&p-Xylenes
2-Methylpentane
i-Butene

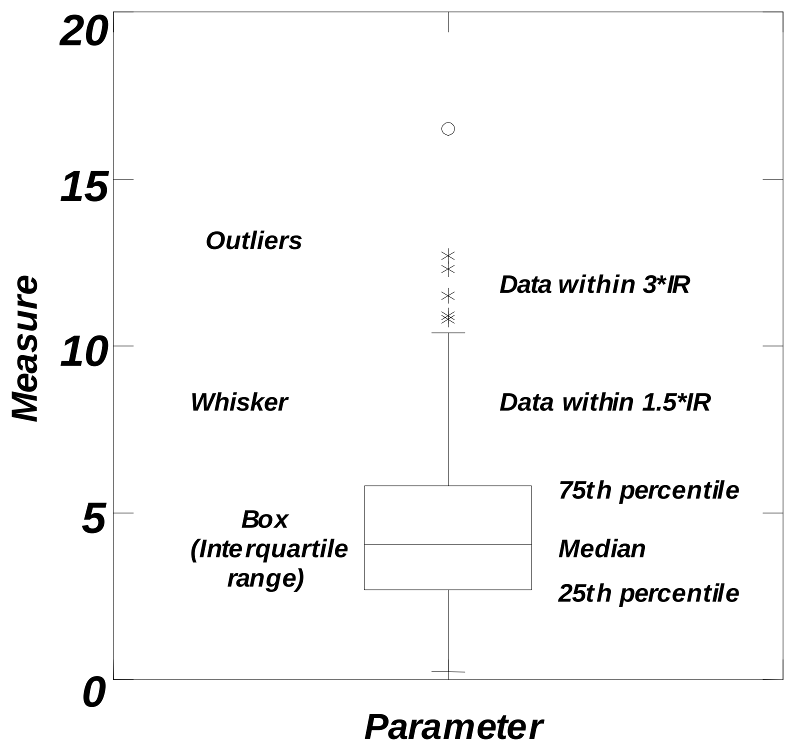
Main et al., 1999

Determine the median value of all species concentrations over the desired time frame (e.g., sample time, month, year) and then sort the median values.

Understanding a Box Plot

- Box plots are commonly used to display a large amount of data and are particularly useful in assessing differences between data.

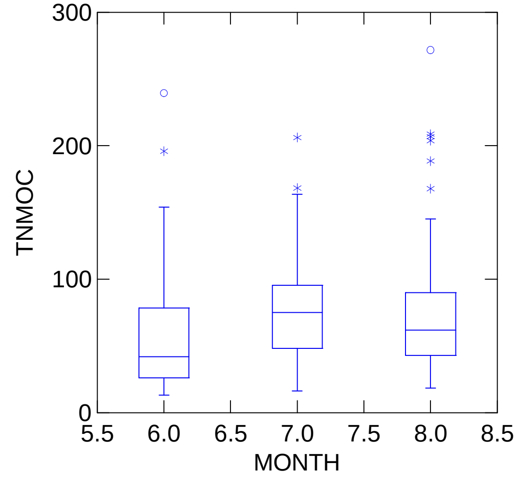
- Box plots are drawn in different ways by different programs. However, most box plots show an interquartile range and some way to illustrate data outside this range. Analysts should understand the definitions of their drawing packages.

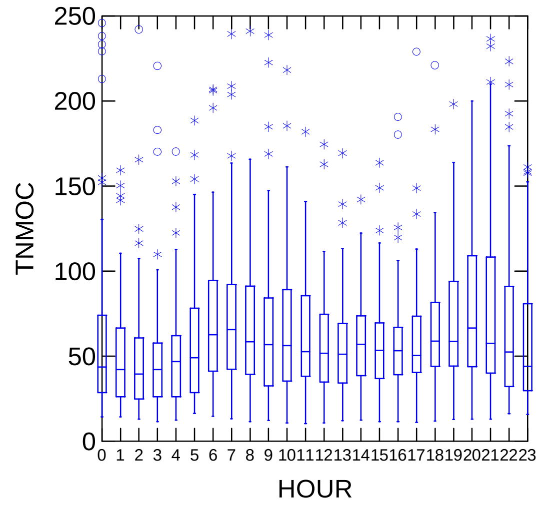


This figure illustrates how SYSTAT software defines a box plot.

Variation of Species by Month and Time of Day (1 of 2)

- This example shows how TNMOC concentrations (in ppbC) vary by month and by time of day at an urban PAMS site.
- The TNMOC concentrations did not vary significantly by month over the PAMS season at this site (note the large overlap among the interquartile ranges).
- TNMOC concentrations are higher in the morning commute time and again later in the evening consistent with traffic patterns near the site.

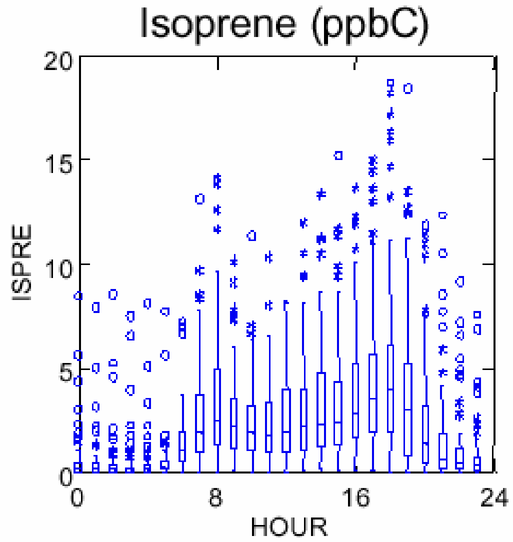


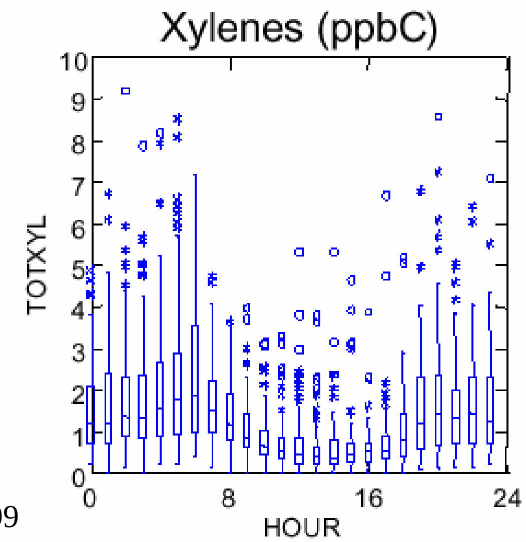


These plots were prepared using SYSTAT.

Variation of Species by Month and Time of Day (2 of 2)

- This example illustrates the diurnal variation of isoprene and the xylenes at a Type 1/4 site in 1997 (Lums Pond, DE).
- Isoprene emissions are a function of sunlight and temperature and thus we expect higher concentrations during the daylight hours.
- The xylenes, key components of motor vehicle exhaust, show a marked decrease at midday, probably indicative of a more aged air mass impacting the site.

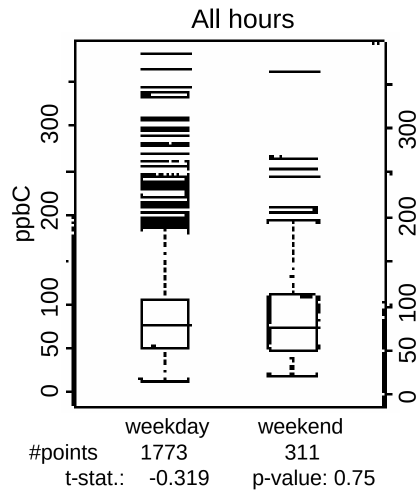




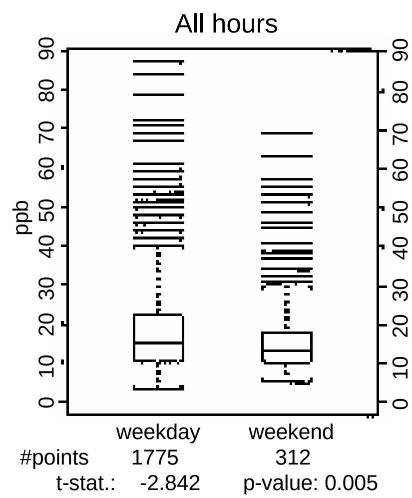
Box plots prepared using SYSTAT. Main et al., 1999

Variation of Species by Day of Week (1 of 2)

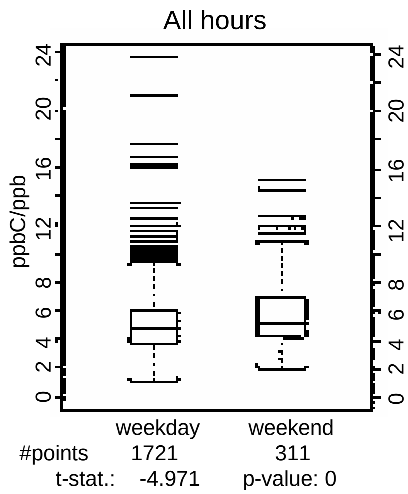
- Comparing concentration data by day of week can be useful to identify differences in emission patterns and can provide clues about the impact of precursor emissions on ozone levels.
- This example shows the weekend vs. weekday distributions of TNMOC (labeled TNMHC in the figures), NO_x, and the TNMOC/NO_x ratio at an urban PAMS site.
- Results of two-sided t-test for the equality of the means between weekdays and weekends are shown at the bottom of each plot. A negative "t-stat" indicates that the weekday mean is less than the weekend mean. The significance level of the test is indicated by the "p-value"; a value of less ≤ 0.01 indicates that the difference in the means is statistically significant at the 99% level.



TNMHC



NO_x



TNMHC/NO_x ratio

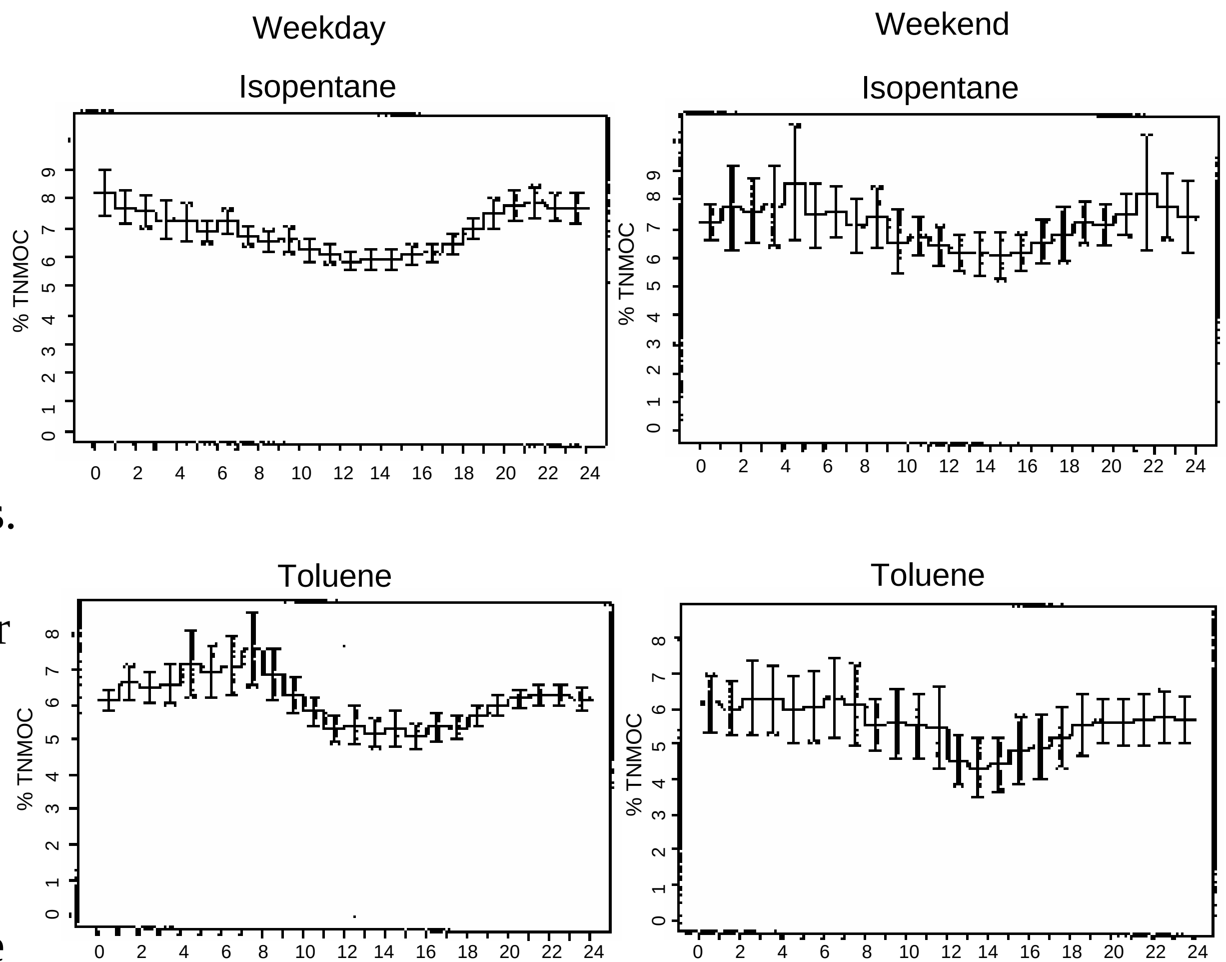
Plots prepared using AMDAS adapted from Stoeckenius et al., 1998

Variation of Species by Day of Week (2 of 2)

- This example shows the weekend vs. weekday distributions of isopentane and toluene weight percents by time of day at an urban PAMS site.

- Error bars on these figures represent 90% confidence intervals for the hourly means. In general, confidence intervals are larger for smaller sample size (thus smaller for weekend vs. weekday plots).

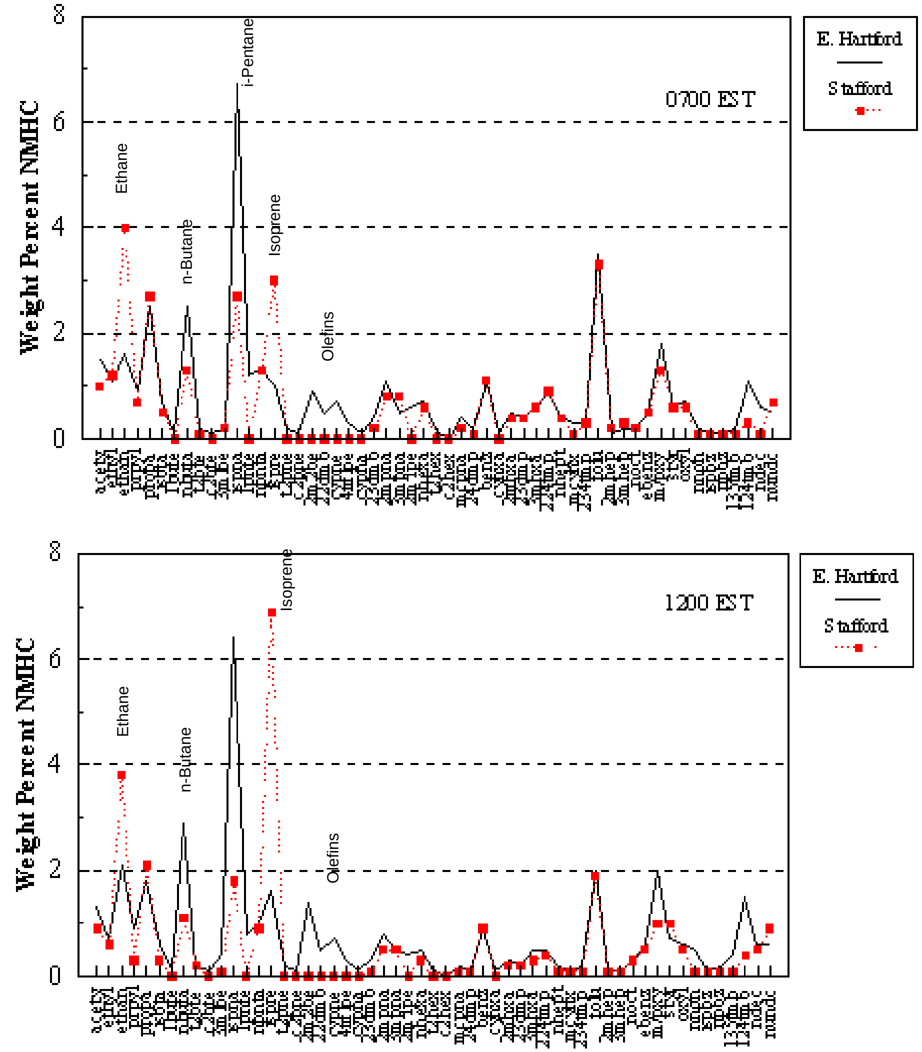
- The toluene weight fraction appears to show a more discernible morning commute time peak on the weekdays than on the weekends.



Plots prepared using AMDAS from Stoeckenius et al., 1998

Comparing Composition Among Sites

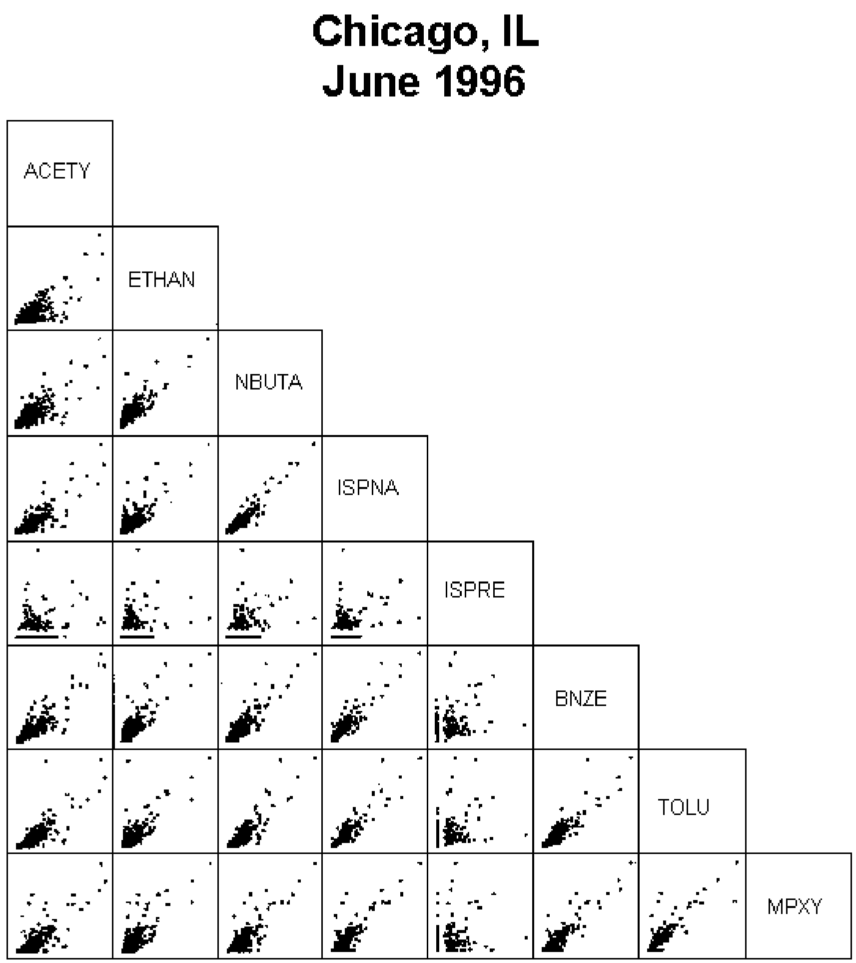
- Differences in composition among sites can reveal important information.
- This example shows average fingerprints at E. Hartford (Type 2) and Stafford (Type 3), CT for June 1995 at 0700 and 1200 EST. The compositions are similar at the two sites with the exceptions of the greater abundance of ethane (relatively unreactive) and isoprene (biogenic) at Stafford, and the greater abundance of several olefins at E. Hartford.



Assessing Relationships Among Species

- Example scatter plot matrix (SPLOM) used to assess relationships among hydrocarbons. To interpret a SPLOM, locate where a row and column intersect (e.g., ACETY-acetylene and MPXY-m-&p-xylenes on the bottom left hand corner). The intersection is the scatter plot of the row variable on the vertical axis against the column variable on the horizontal axis. Each column and row are scaled so that data points fill each frame.

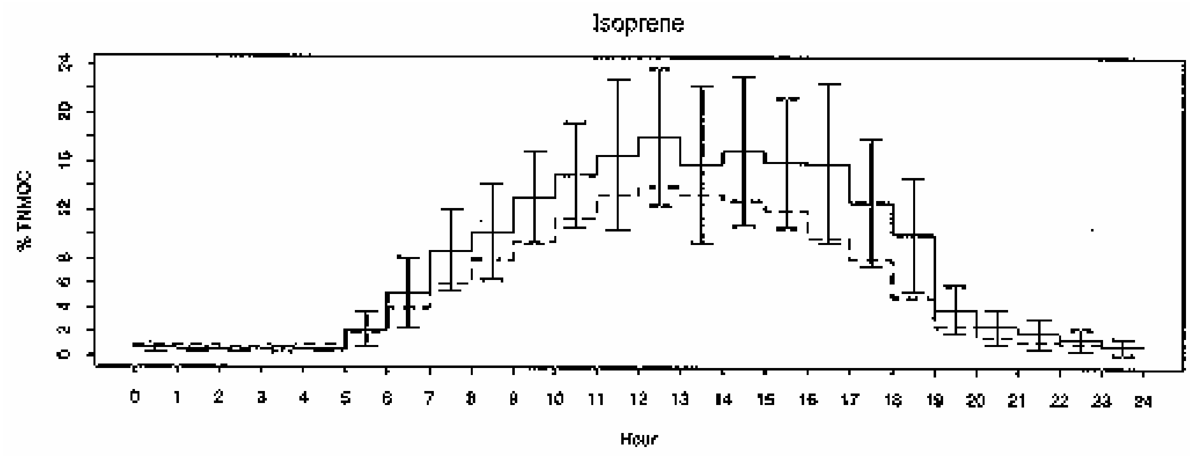
- In this example, the isoprene (ISPRE) data do not appear to correlate well with the other hydrocarbons shown. In contrast, n-butane (NBUTA) and i-pentane (ISPNA) correlate very well implying that the two hydrocarbons are from similar sources.

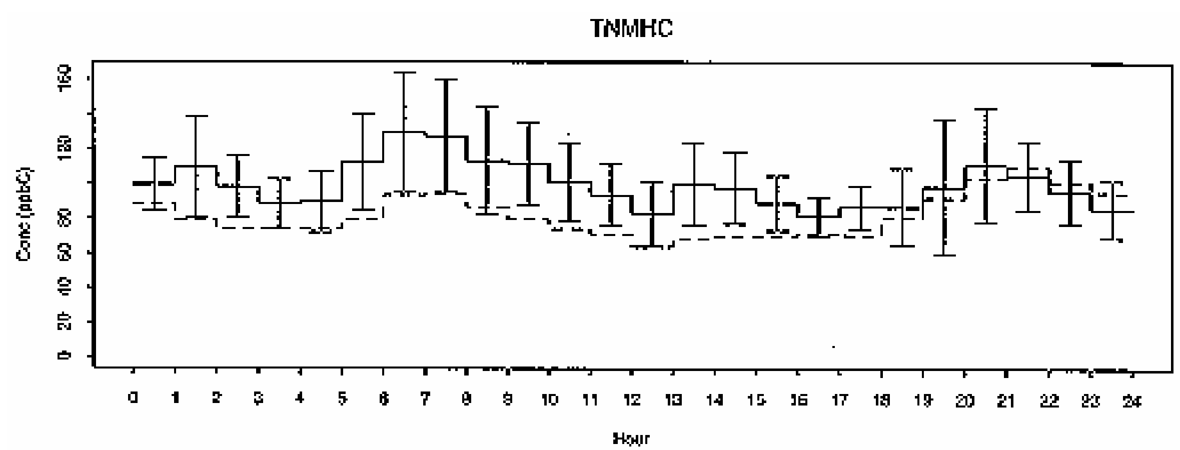


Scatter plot matrix of eight abundant hydrocarbons at Chicago, IL during 1996. Prepared using SYSTAT.

Comparing PAMS Data on Ozone Episode and Non-episode Days (1 of 3)

- To understand potentially unique characteristics of high ozone concentration events (episodes), the analyst should compare VOC composition on episode vs. non-episode days.
- In this example, a 90% confidence interval for the episode day means is provided. One would expect the confidence interval to be smaller for the non-episode days as the sample size is much larger.
- The weight fraction of isoprene appears to be higher on episode days than on non-episode days in this example possibly reflecting higher temperatures characteristic of episode conditions.





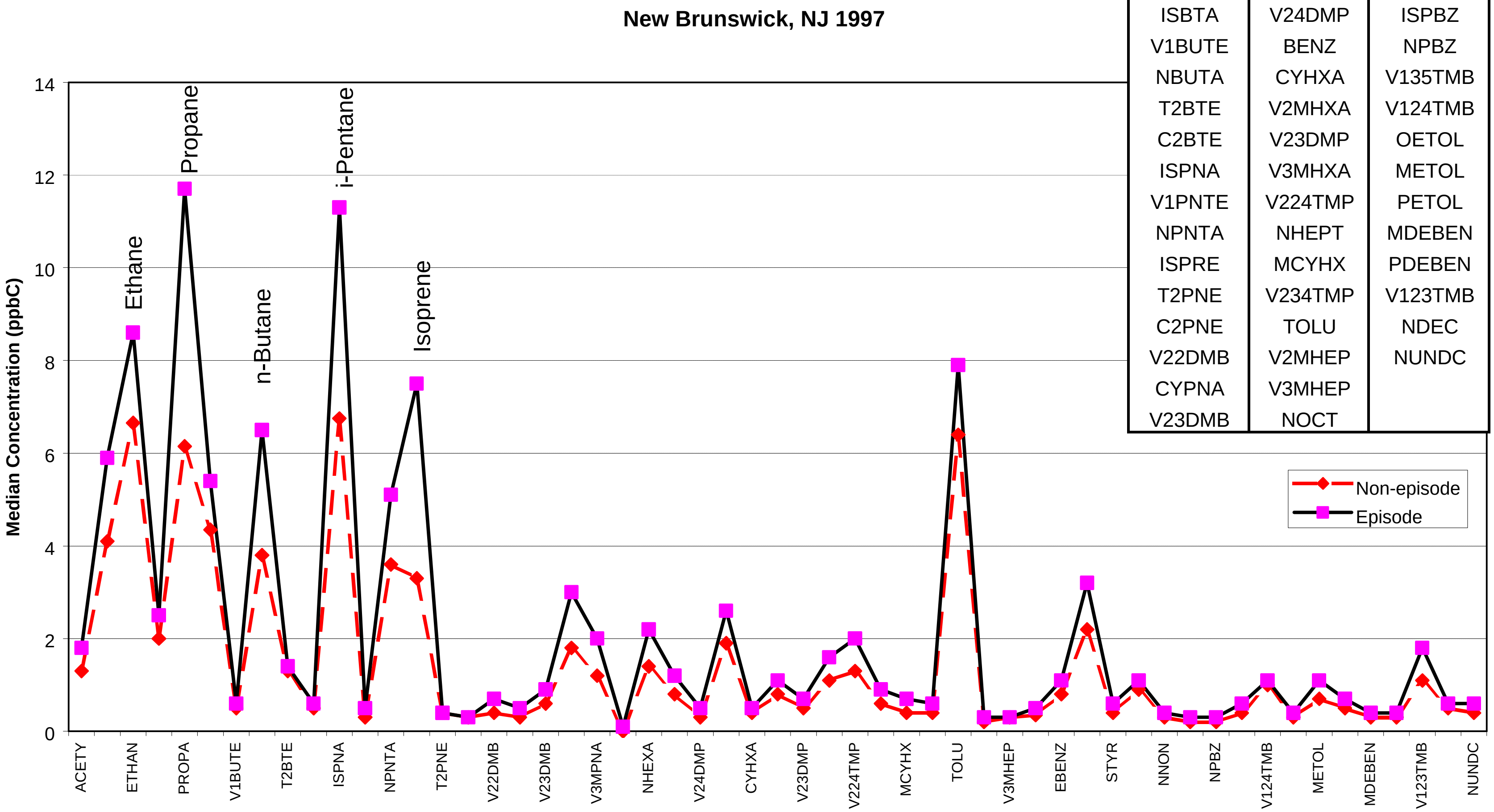
— Episode day
- - Non-episode day

Plots prepared using AMDAS from Stoeckenius et al., 1998

Comparing PAMS Data on Ozone Episode and Non-episode Days (2 of 3)

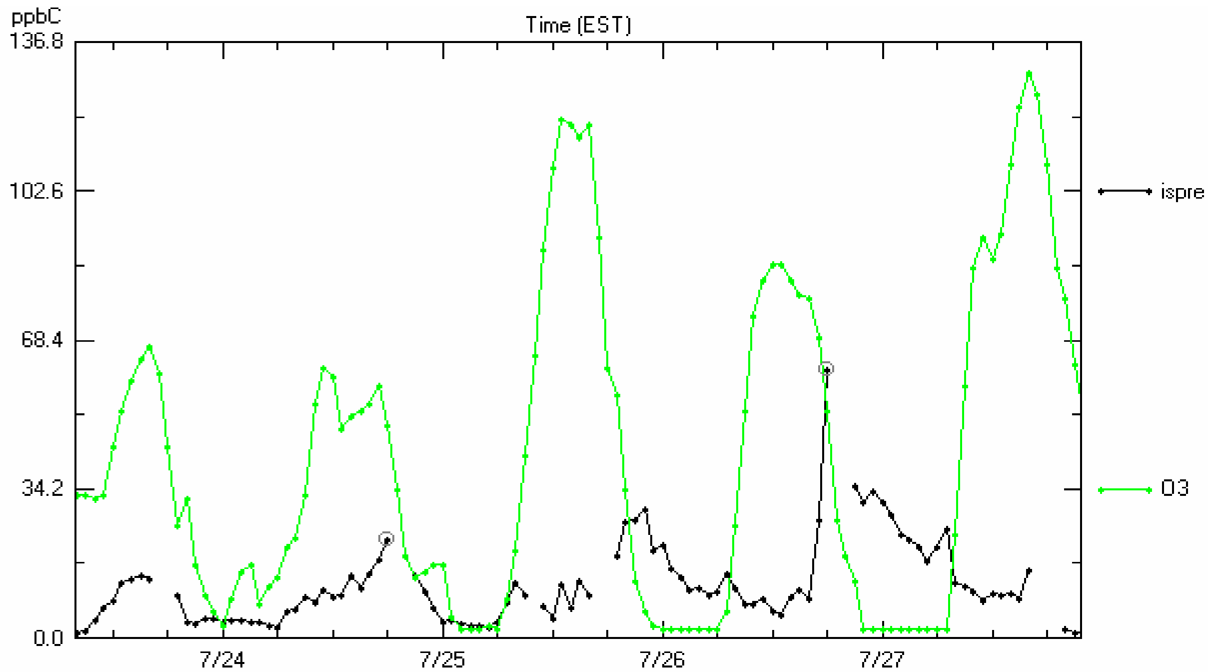
COMPLETE LIST OF SPECIES ABBREVIATIONS

ACETY	V2MPNA	EBENZ
ETHYL	V3MPNA	M_PXY
ETHAN	V2M1PE	STYR
PRPYL	NHEXA	OXYL
PROPA	MCPNA	NNON
ISBTA	V24DMP	ISPBZ
V1BUTE	BENZ	NPBZ
NBUTA	CYHXA	V135TMB
T2BTE	V2MHXA	V124TMB
C2BTE	V23DMP	OETOL
ISPNA	V3MHXA	METOL
V1PNTE	V224TMP	PETOL
NPNTA	NHEPT	MDEBEN
ISPRE	MCYHX	PDEBEN
T2PNE	V234TMP	V123TMB
C2PNE	TOLU	NDEC
V22DMB	V2MHEP	NUNDC
CYPNA	V3MHEP	
V23DMB	NOCT	



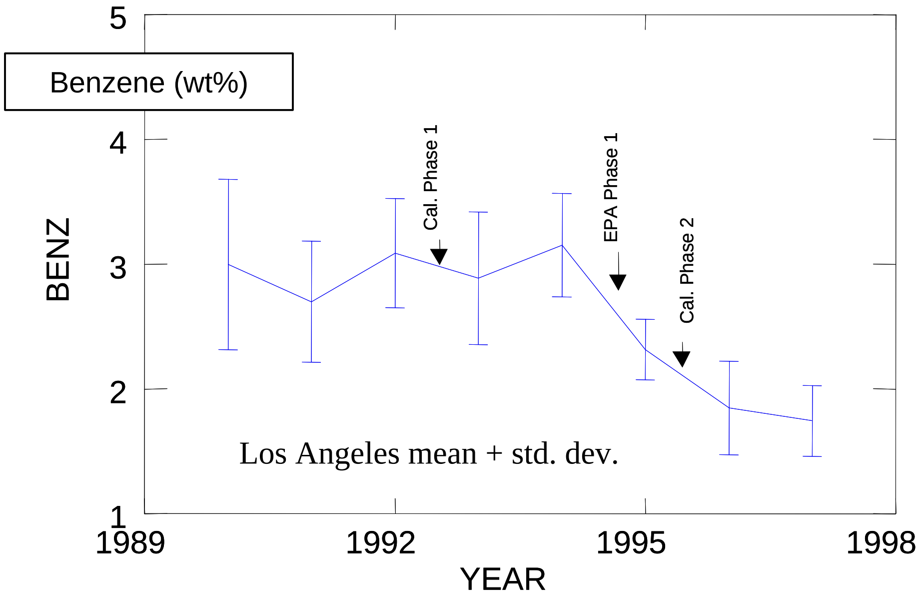
Median concentrations at New Brunswick, NJ in 1997 from 0600 to 0800 EST on episode days (peak ozone ≥ 100 ppb) and non-episode days. The higher concentrations of the least reactive paraffin species (i.e., ethane, propane, butanes) on episode days may indicate a build up of VOCs overnight. The higher isoprene in the morning may be a function of temperature. (Main et al., 1999)

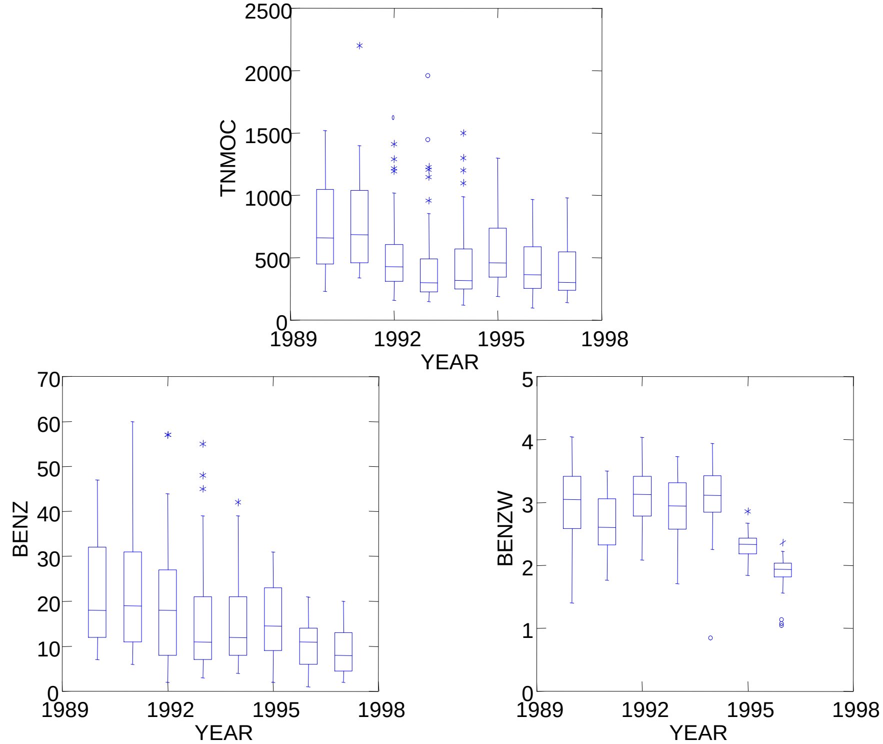
Comparing PAMS Data on Ozone Episode and Non-episode Days (3 of 3)



- Some analysts have found that individual PAMS species may be useful in assisting ozone episode forecasting efforts. Time series, box plots, and fingerprint plots may be useful in assessing this.
- The example shown above is a time series of ozone (O3 in ppb) and isoprene (ISPRE in ppbC) concentrations for a few days in July at a rural Georgia site. The isoprene concentrations appear to be higher and later in the day on the days preceding high ozone concentrations. Higher isoprene concentrations overnight may also be an indicator of conditions the next day that are conducive for ozone formation.

Tracking Changes in PAMS Species (1 of 2)





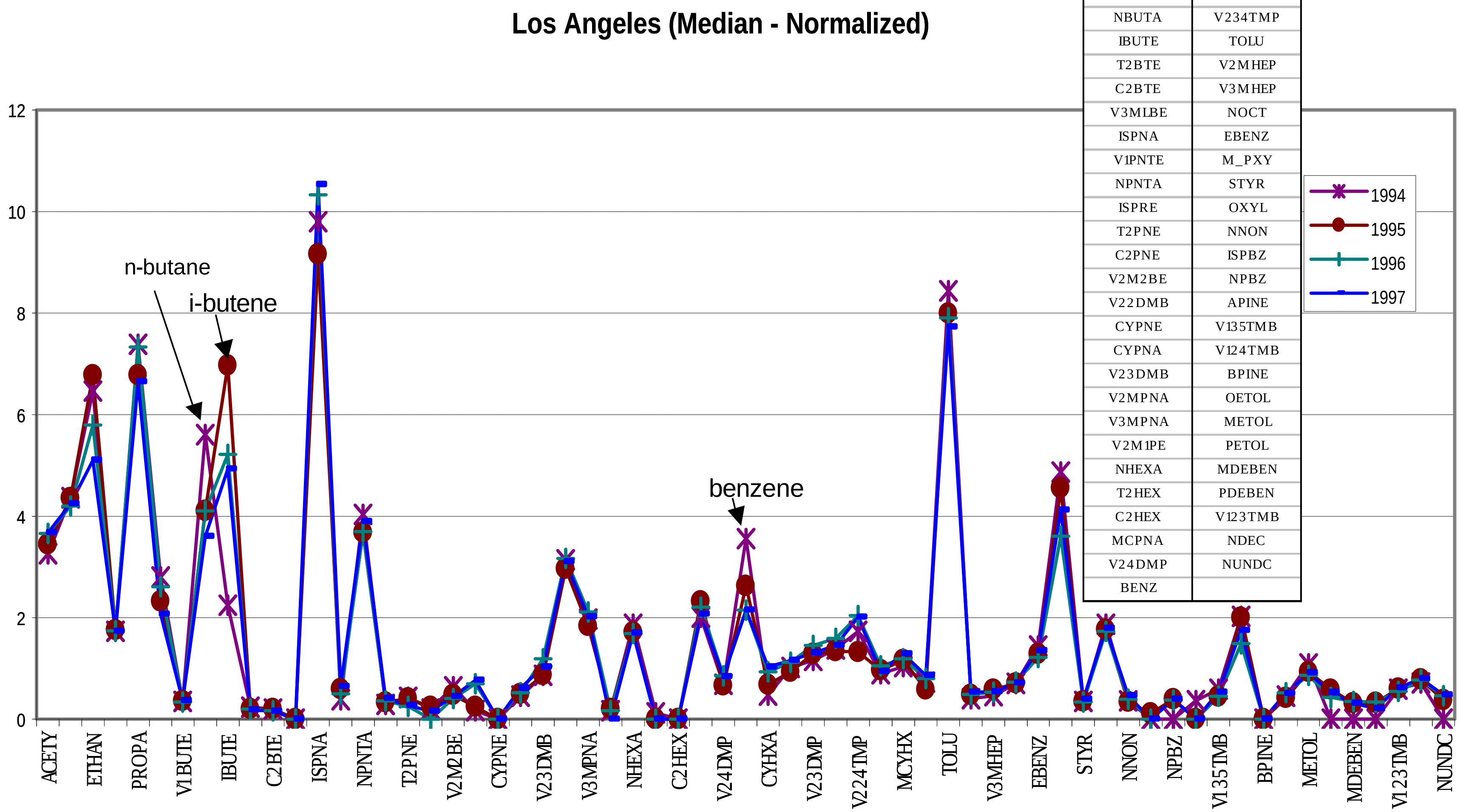
Main et al., 1999

- The federal reformulated gasoline (RFG) implementation occurred in early 1995; benzene fuel levels were specifically targeted for reduction (EPA Phase 1). In addition, California had separate fuel requirements that further reduced fuel volatility (Cal. Phase 1) and benzene levels (Cal. Phase 2) as shown in the left-hand figure.
- The figures at the right show annual box plots of ambient TNMOC (ppbC), benzene concentration (BENZ in ppbC), and benzene weight percent (BENZW) in Los Angeles. The drop in ambient benzene weight percent between 1994 and 1995 is dramatic. Additionally, fuel benzene measurements also declined between 1994 and 1995. It is important to attempt to explain the changes observed in the ambient air with the control actions.

Tracking Changes in PAMS Species (2 of 2)

ACETY	CYHXA
ETHYL	V2MHXA
ETHAN	V23DMP
PRPYL	V3MHXA
PROPA	V224TMP
ISBTA	NHEPT
V1BUTE	MCYHX
NBUTA	V234TMP
IBUTE	TOLU
T2BTE	V2MHEP
C2BTE	V3MHEP
V3MLBE	NOCT
ISPNA	EBENZ
V1PNTE	M_PXY
NPNTA	STYR
ISPRE	OXYL
T2PNE	NNON
C2PNE	ISPBZ
V2M2BE	NPBZ
V22DMB	APINE
CYPNE	V135TMB
CYPNA	V124TMB
V23DMB	BPINE
V2MPNA	OETOL
V3MPNA	METOL
V2M1PE	PETOL
NHEXA	MDEBEN
T2HEX	PDEBEN
C2HEX	V123TMB
MCPNA	NDEC
V24DMP	NUNDC
BENZ	

COMPLETE LIST OF SPECIES ABBREVIATIONS



This figure shows the median composition at Los Angeles from 1994 through 1997 normalized over the identified hydrocarbons (Main et al., 1999). The benzene and n-butane fractions significantly decreased after 1995. These changes are consistent with the fuel changes to reduce benzene, aromatics, and fuel volatility. The i-butene fraction increased. This hydrocarbon is a thermal decomposition product of methyl-tert-butyl-ether (MTBE); MTBE is a species that was added to RFG to meet oxygen requirements.

Summary

- After data validation, numerous investigative analyses should be made of the PAMS VOC data. To meet PAMS data analysis objectives and attempt to answer related questions, several analyses are shown in this and other sections.
- Spatial and temporal characteristics of the data are explored using summary statistics, box plots, scatterplots, time series, and fingerprint plots. Useful tools for these analyses include AMDAS, spreadsheets, and other statistical packages.

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